

# Influence of interphase anisotropy on lamellar eutectic growth patterns

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## Abstract

It is well documented in many experiments that crystallographic effects play an important role in the generation of two-phase patterns during the solidification of eutectic alloys. In particular, in lamellar composites, large patches of perfectly aligned lamellae are frequently observed. Moreover, the growth direction of the lamellae often markedly differs from the direction of the temperature gradient (the lamellae are tilted with respect to the main growth direction). Both of these effects cannot be explained either by the standard theory or the available numerical models of eutectic growth, which all assume the interfaces to be isotropic. We have developed a phase-field model in which the anisotropy of each interface (solid-liquid and solid-solid) can be separately controlled, and we have investigated the effect of interface anisotropy on the growth dynamics. We have found that anisotropy of the solid-solid interphase boundary free energy dramatically alters the growth dynamics. Tilted lamellae result from the modified equilibrium condition at the triple lines, in good agreement with a theoretical conjecture proposed recently. In three dimensions, the interphase boundaries tend to align with directions of minimal energy. We have also performed simulations in which two grains with different anisotropies are in competition. In all cases, the grain containing the boundaries with the lowest energies was selected after a transient. These results shed new light on the selection of growth patterns in eutectic solidification.

## I. INTRODUCTION

During directional solidification, a sample is pulled with a constant velocity  $V$  in a fixed thermal gradient  $G$  aligned with the  $z$  direction. For a non-faceted binary eutectic alloy, this leads to the formation of composite materials: the solid consists of two phases  $\alpha$  and  $\beta$ , which grow together by diffusive exchange of components through the liquid [1]. In the absence of interfacial anisotropy, the solid phases grow next to each other, the interphase boundaries (interfaces between  $\alpha$  and  $\beta$ ) are aligned with the growth direction, and Young’s law is satisfied at the trijunction points. Here, we investigate several phenomena that arise when the interphase boundaries (IB) are anisotropic.

Whereas, in non-faceted substances, the solid-liquid surface free energy is only weakly anisotropic, the anisotropy of solid-solid IB can be strong. This anisotropy depends on the relative orientation of the two solids with respect to each other. The anisotropy function thus is not an intrinsic property of the material, but differs between different *eutectic grains* [2] – that is, portions of the solid in which the orientations of the two phases remain homogeneous. One may distinguish *floating grains* with low anisotropy from *locked grains* with high anisotropy [3]. In the latter, the IB may remain “locked” onto a direction of low energy, irrespective of the orientation of the grain with respect to the temperature gradient.

For our studies, we have used the grand-canonical phase-field model described in Refs. [4, 5]. In this model, which provides quantitative results due to a well-controlled thin-interface limit [4], the anisotropy of each interface can be chosen independently [5]. We write the surface energy  $\gamma_{\alpha\beta}$  as

$$\gamma_{\alpha\beta} = \bar{\gamma}_{\alpha\beta} a_c(\theta, \phi), \quad (1)$$

where  $\bar{\gamma}_{\alpha\beta}$  is a constant which we take equal to the solid-liquid interface free energies, and  $a_c(\theta, \phi)$  is the dimensionless anisotropy function. Here,  $\theta$  and  $\phi$  are the standard polar angles with respect to the  $z$  and  $x$  axis, respectively, and we take the convention that  $\theta = \phi = 0$  corresponds to a minimum of  $a_c$  (that is, a minimum in the interface energy occurs when the boundary is in the  $xz$  plane). In the so-called rotating directional solidification set up [6], the orientation of the eutectic crystal with respect to the temperature gradient can be

changed during the experiment. We describe such rotations around the  $y$  and  $z$  axes by the angles  $\theta_R$  and  $\phi_R$ , respectively.

We consider a generic binary eutectic alloy of symmetric phase diagram, with equal volume fractions of the two solids. In this situation, the capillary and thermal lengths associated with the two solids are equal.

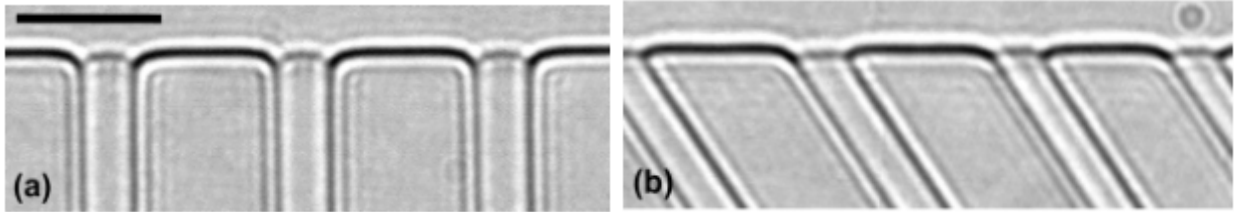


FIG. 1: Snapshots of directional solidification in thin samples of the transparent alloy  $\text{CBr}_4\text{-C}_2\text{Cl}_6$  (adapted from Akamatsu et al. [7]); Bar:  $20\ \mu\text{m}$ .

## II. TILTED LAMELLAE

As shown in Fig. 1b, lamellae do not always grow aligned with the temperature gradient (which is vertical in the figure). The growth angle results from a competition between the external temperature field and the interface anisotropy. Recently, a conjecture was made that provides a prediction for the growth angle [7]. It is based on the observation (see Fig. 1) that the solid-liquid interfaces (the “lamellae heads”) remain symmetric even when the lamellae are tilted. This implies that the Cahn-Hoffman surface tension vector [8] must be aligned with the  $z$  axis (see Refs. [5, 7] for details). From this hypothesis, one obtains

$$\gamma_{\alpha\beta}(\theta - \theta_R) \sin \theta + \gamma'_{\alpha\beta}(\theta - \theta_R) \cos \theta = 0, \quad (2)$$

where the prime denotes differentiation with respect to  $\theta$ . For a fixed orientation  $\theta_R$  of the eutectic grain, this is a nonlinear equation for the interface orientation  $\theta$ , which can easily be solved numerically for arbitrary anisotropy functions  $a_c(\theta)$ . As long as the interface stiffness  $\gamma + \gamma''$  is positive for all angles, this equation has a unique solution. For negative stiffness, there are ranges of  $\theta_R$  for which there exist three solutions, of which one corresponds to an

orientation that is present on the equilibrium shape (stable solution), one to an unstable orientation, and the third to a metastable orientation. This is the prediction against which we will compare our numerical results.

We have performed simulations in two dimensions for various anisotropy functions, with the results shown in Fig. 2. We have used smooth functions such as the standard four-fold anisotropy ( $a_c(\theta) = 1 - \epsilon_4 \cos(4[\theta - \theta_R])$ ), as well as a function with a deep but smooth Gaussian minimum,  $a_c(\theta) = 1 - \epsilon_c \exp((\theta - \theta_R)^2/w_c^2)$  with  $\epsilon$  being the magnitude of the anisotropy and  $w_c$  the width of the minimum. Note that, for these anisotropy functions, depending on the value of  $\epsilon$ , missing orientations (signifying multiple solutions for a particular rotation angle) can occur.

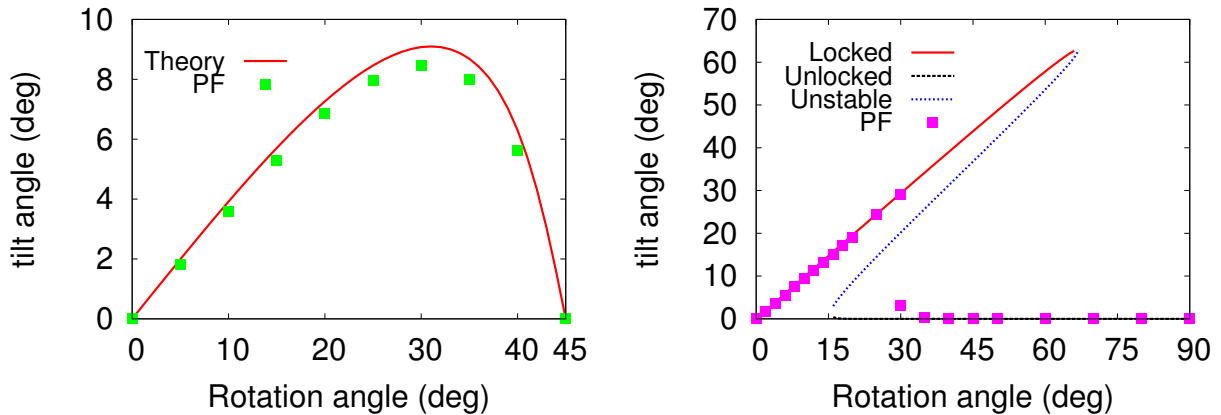


FIG. 2: Interphase tilt angle for (left) smooth anisotropy function ( $\epsilon_4 = 4\%$ ) (right) cusp anisotropy function with  $\epsilon_c = 20\%$ . The solid lines indicate the tilt angle predicted by Eq. 2; squares denotes the phase-field (PF) results.

For smooth anisotropies, our results follow quite nicely the prediction of the symmetric pattern approximation. When multi-valued solutions exist for a particular rotation angle – one solution for a locked branch, one for an unlocked (zero angle) branch and one for an unstable branch (dotted in Fig. 2) – the locked branch is well reproduced up to a certain angle ( $\approx 30$  deg). For rotation angles higher than this value, simulations always end up on the unlocked branch. When the rotation angle is slowly increased or decreased in small steps, the “jump” between the two branches always occurs at the same angle. Therefore, multiple solutions for a given rotation angle are never observed. This is in contrast to results obtained

with the boundary integral (BI) method [5]. The reason for this discrepancy between the models is as of yet unknown.

### III. GRAIN COMPETITION

For investigations of anisotropy in the azimuthal plane (the plane perpendicular to the temperature gradient), three-dimensional simulations are necessary. In bulk lamellar eutectics there are complex interactions between multiple eutectic grains and interfaces. In the absence of anisotropy, the microstructure consists of random lamellar patterns, because no particular orientation is favoured. When anisotropy is imparted in the system, the system begins to order depending on the underlying crystal structure of the solid. For example, if a two-fold anisotropy is present in the system, we obtain a perfectly lamellar array after a short transient. The lamellae are always aligned with a minimum in the interface energy function. When the latter is rotated with respect to the lateral “walls” (described by no-flux boundary conditions), the lamellae are perpendicular to the boundaries close to the walls, but turn to align with the minimum-energy direction over a distance of less than one lamellar spacing.

In order to investigate the competition between eutectic grains, we have considered the growth of two different eutectic grains with different anisotropy functions  $a_c$ . In the example shown in Fig. 3, the interface energy between the “red” and “green” solids is given by  $a_c = 1.0 + 0.3 \cos[2(\phi - \phi_R)]$ , whereas all other IB’s are isotropic ( $a_c = 1$ ). The simulation is started from a random tiling of the system with the various phases. A random lamellar pattern develops in the beginning, with a presence of local order in the system while there is no global order (Fig. 3b). Later on, lamellae of the red-green composite start to grow along the preferred orientation. Finally, the system is left with only anisotropic interfaces, which are aligned with a minimum energy direction (Fig. 3c). In essence, surface tension anisotropy along the interphase boundaries induces an orientation relationship between the anisotropic phases yielding a regular lamellar array from an irregular eutectic structure.

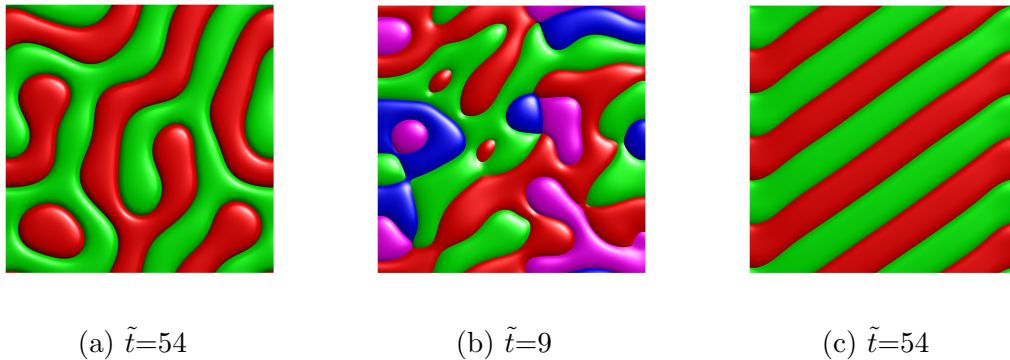


FIG. 3: Top view of three-dimensional simulations. (a) Random lamellar pattern formation in a single eutectic grain in the absence of anisotropy. (b and c) time evolution of two eutectic grains: colors of  $\alpha_1$ ,  $\beta_1$ ,  $\alpha_2$  and  $\beta_2$  are red, green, blue and magenta, respectively. The IB between red and green phases are anisotropic, all others isotropic. The dimensionless time is  $\tilde{t} = tV^2/D$

#### IV. CONCLUSIONS

We have used phase-field simulations to investigate the influence of crystallographic effects on eutectic solidification patterns. We have found that an anisotropy of the solid-solid inter-phase boundaries can lead to important departures from the behavior known for eutectics with isotropic interfaces. For an anisotropy in a plane parallel to the temperature gradient, the competition between the external temperature gradient and the IB energy selects a growth direction that depends on the anisotropy strength and differs from the direction of the temperature gradient, leading to the growth of tilted lamellae. Anisotropy in the plane perpendicular to the temperature gradient (azimuthal plane) leads to a selection of particular orientations of the lamellae, which correspond to minimum energy directions. This obviously favors the emergence of ordered (parallel) lamellar arrays. A highly interesting perspective is to understand this selection more quantitatively, and to relate phase-field simulations to experiments on Al-Cu alloys in which the crystallographic effects were characterized in detail [9, 10].

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